

Thermoluminescence of Simulated Interstellar Matter after Gamma-ray Irradiation

Forsterite, Enstatite and Carbonates

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Abstract. Interstellar matter is known to be strongly irradiated by radiation and several types of cosmic ray particles. Simulated interstellar matter, such as forsterite Mg_2SiO_4 , enstatite MgSiO_3 and magnesite MgCO_3 has been irradiated with the ^{60}Co gamma-rays in liquid nitrogen, and also irradiated with fast neutrons at 10 K and 70 K by making use of the low-temperature irradiation facility of Kyoto University Reactor (KUR-LTL. Maximum fast neutron dose is $10^{17} n_f/\text{cm}^2$). After irradiation, samples are stored in liquid nitrogen for several months to allow the decay of induced radioactivity. We measured the luminescence spectra of the gamma ray irradiated samples during warming to 370K using a spectrophotometer. For the forsterite and magnesite, the spectra exhibit a rather intense peak at about 645 – 655 nm and 660 nm respectively, whereas luminescence scarcely appeared in olivine sample. The spectra of forsterite is very similar to the ERE of the Red Rectangle.

Key words. forsterite– irradiation– gamma-ray

1. Introduction

Interstellar matter will be irradiated by electromagnetic energy and several cosmic ray particles, such as gamma rays, neutrons, protons and heavy-ions etc. Irradiation will cause some changes in these materials, especially to their optical properties. Investigation of this problem is expected to advance our understanding of interstellar and circumstellar matter. Especially, it is well known that extremely large fluxes of neutrons and gamma-rays have been emitted during super-nova explosions and during the so called Hayashi-phase in the early stage of protostellar systems. Moreover, interplanetary-dust is often irradiated by gamma rays and fast neutrons during the periods of flare activity on the sun, which is always repeated. Interstellar and circumstellar space is typically at extremely low temperature and is always irradiated over cosmological time-scale.

Though little is known about irradiation environment outside of our solar system, it is natural to suppose that there are the regions with sufficient strength of irradiation to cause thermoluminescences, for example, a region

of not so extremely far from a super nova explosion or not so far from a source of gamma ray emission, and/or long time irradiation at extremely low temperature. The effect of this radiation will accumulate in the low temperature environment. It will be observed provided that the condition to release the accumulated energy is realized in circumstellar space.

The effects of irradiation on simple single crystals such as SiO_2 , CaCO_3 and CaF_2 have been investigated by several authors (Nakagawa et al. 1988, Nakagawa et al. 1999) from the viewpoint of solid state physics or material science. However, irradiation of materials of interstellar matter has not been studied. It may then be worthwhile to investigate the effects of irradiation on simulated interstellar matter such as forsterite, enstatite and carbonates. Forsterite and enstatite have been found by many ISO observations in both young and evolved stars and in our own solar system (Waters et al. 1998, Malfait et al. 1998, Wooden et al. 1999). Carbonates such as dolomite ($\text{CaMg}(\text{CO}_3)_2$), breunnerite ($\text{Mg}(\text{Fe}, \text{Mn})(\text{CO}_3)_2$), calcite (CaCO_3), and Mg, Ca-bearing siderite (FeCO_3) were found in CI chondrite (Endress, Zinner and Bischoff 1996). Especially, it should be noted that the broad emission fea-

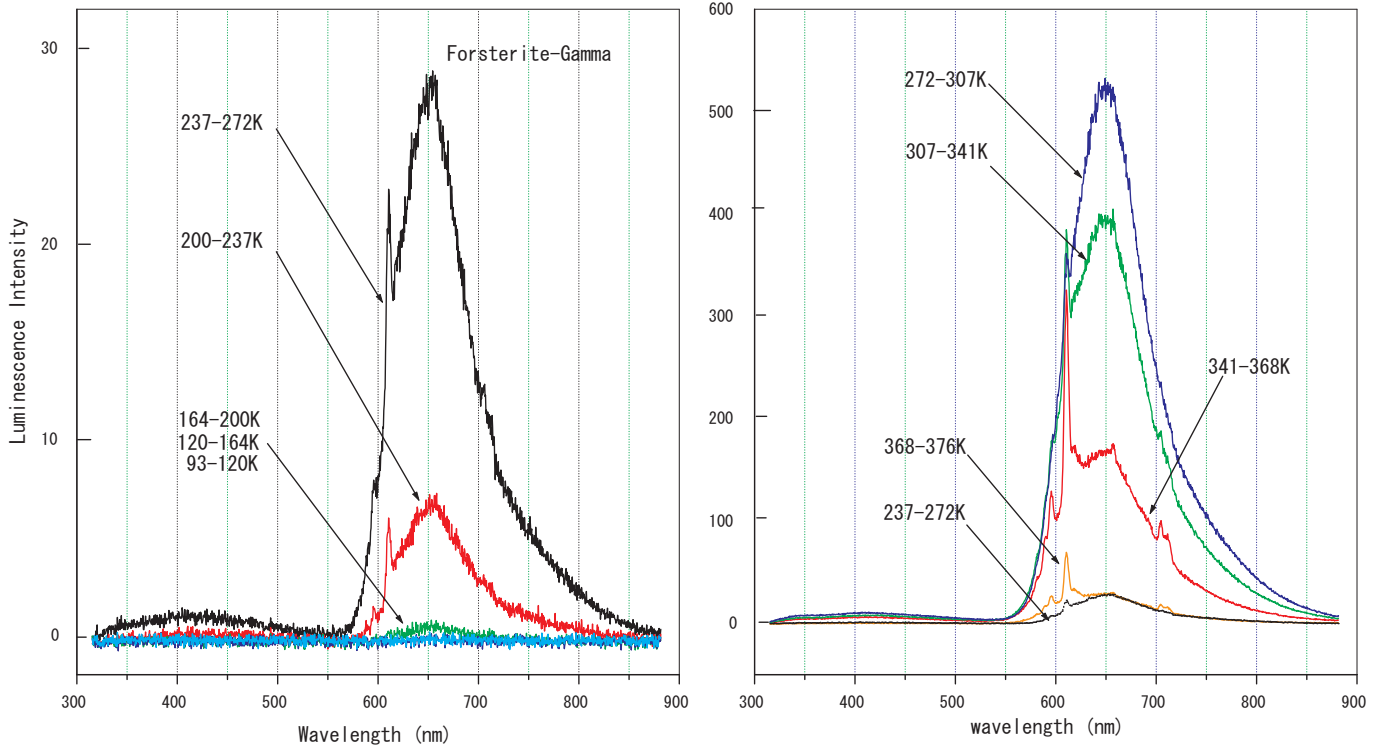


Fig. 1. Thermoluminescence spectra of forsterite (Mg_2SiO_4) single crystals. The left figure corresponds to 93-272K and the right figure 272-376K.

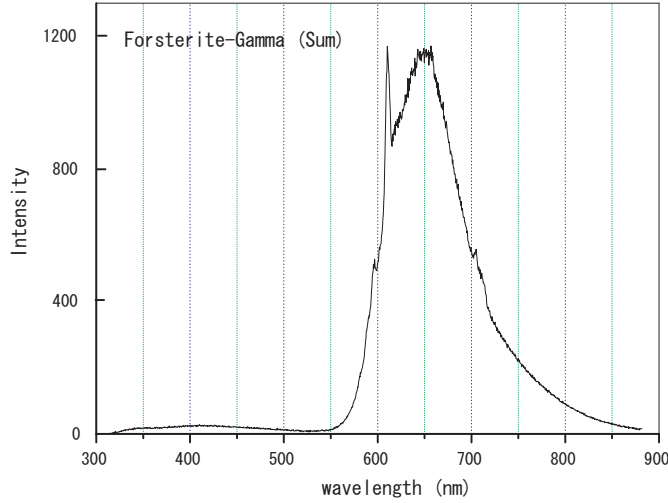


Fig. 2. The sum of total thermoluminescence spectra of forsterite in Fig.1. The peak of the thermoluminescence of Mg_2SiO_4 is about 645-655 nm.

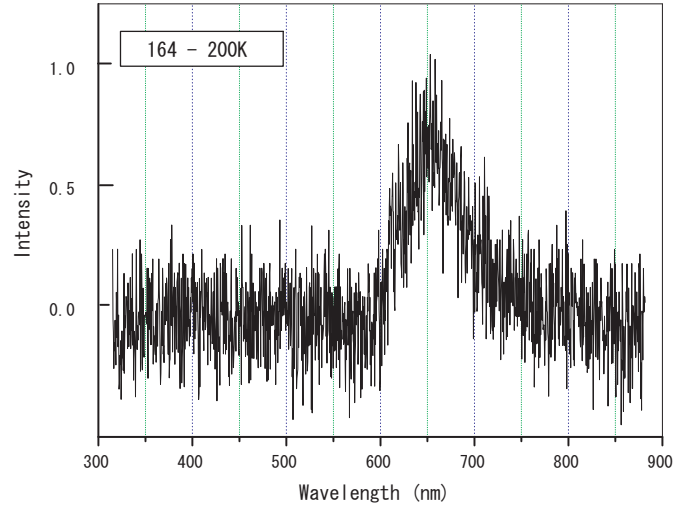


Fig. 3. The earliest (low temperature) thermoluminescence spectra of forsterite at 164-200K. The peak is also at about 645-655 nm.

ture responsible for extended red emission(ERE) appears at about the 500 – 900 nm region in many reflection nebulae, and the Red Rectangle nebula shows sharp emission features over a broad band (Witt and Boroson 1990). In the Red Rectangle nebulae, both PAH-features and crystalline silicates (forsterite and enstatite) were observed (Waters et al., 1998). The effect of irradiation on the optical properties of the simulated interstellar materials such

as forsterite and enstatite by gamma rays and neutrons at low temperature are very interesting.

2. Features of Thermoluminescence spectrum

Bulk samples such as forsterite, Mg_2SiO_4 , natural olivine from Egypt, orthoenstatite MgSiO_3 and natural magnesite MgCO_3 from the Democratic People's Rep. of Korea were irradiated with gamma-rays to a dose of about 10.4×10^4 Gy (J/Kg) in liquid nitrogen using the ^{60}Co gamma-

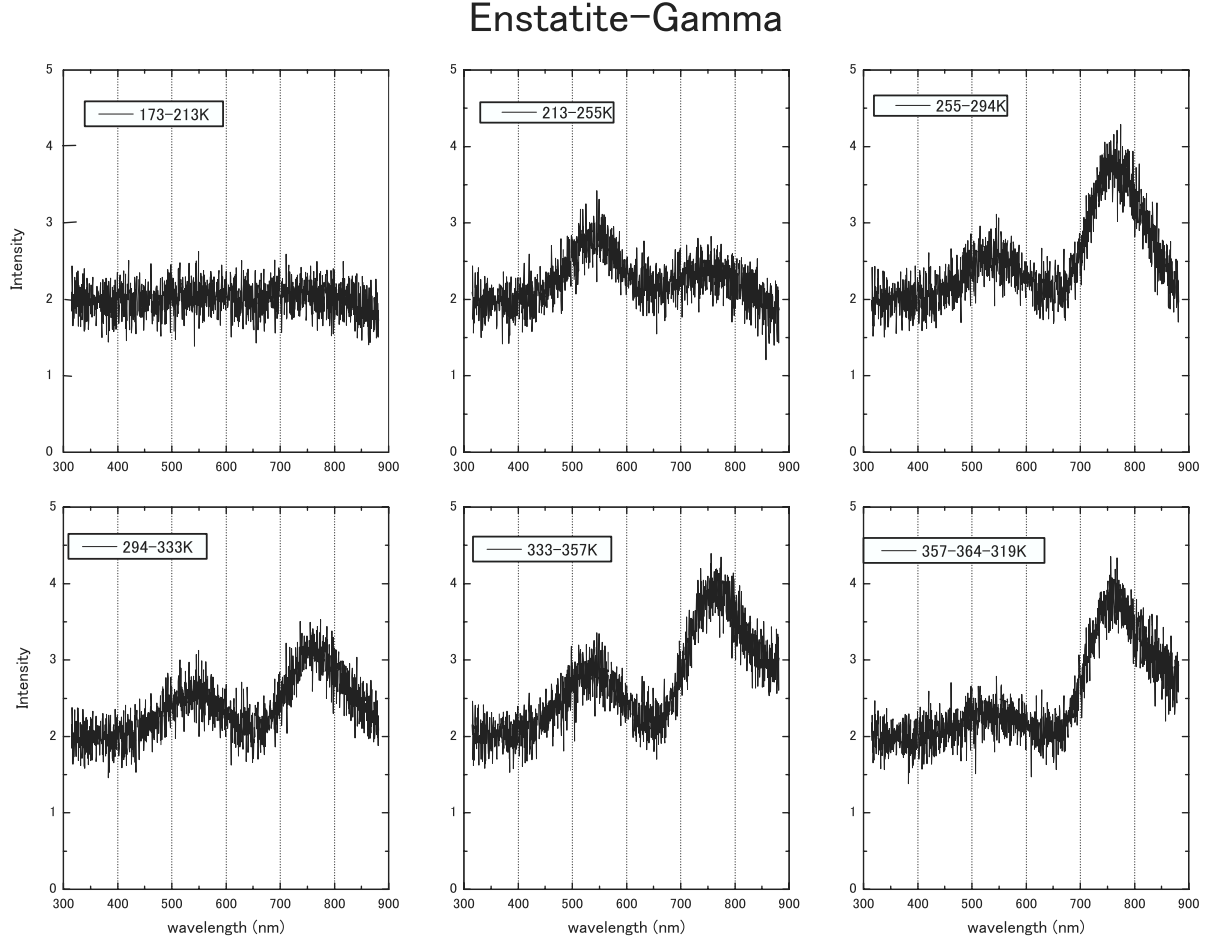


Fig. 4. Thermoluminescence spectra of enstatite (MgSiO_3) single crystals. (The down right-side figure shows the luminescence when the temperature rose from 357K to maximum 364K, and come down till 319K)

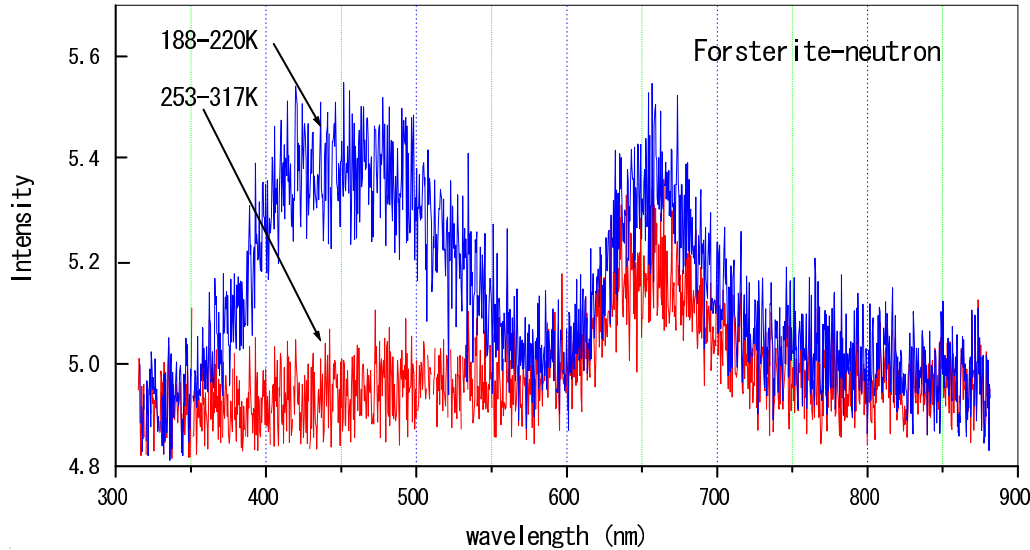


Fig. 5. The thermoluminescences spectra of forsterite after neutron irradiated at 10K.

ray irradiation facility of Kyoto University Reactor. The gamma-rays of ^{60}Co have two peaks at 1.1 MeV and 1.3 MeV. Our samples of forsterite and enstatite were synthesized by Takei and Kobayashi (1974), and Tachibana (2000) using the CZ (Czochralski) and Flux method, respectively with high accuracy. The bulk of the irradiated

forsterite is triangular-shaped (about $6 \times 8 \times 15$ mm size, about 1 mm thickness) and weights about 138 mg. The irradiated enstatite consists of several small fragments (about 1 - 2 mm size) and the total weight is about 114 mg.

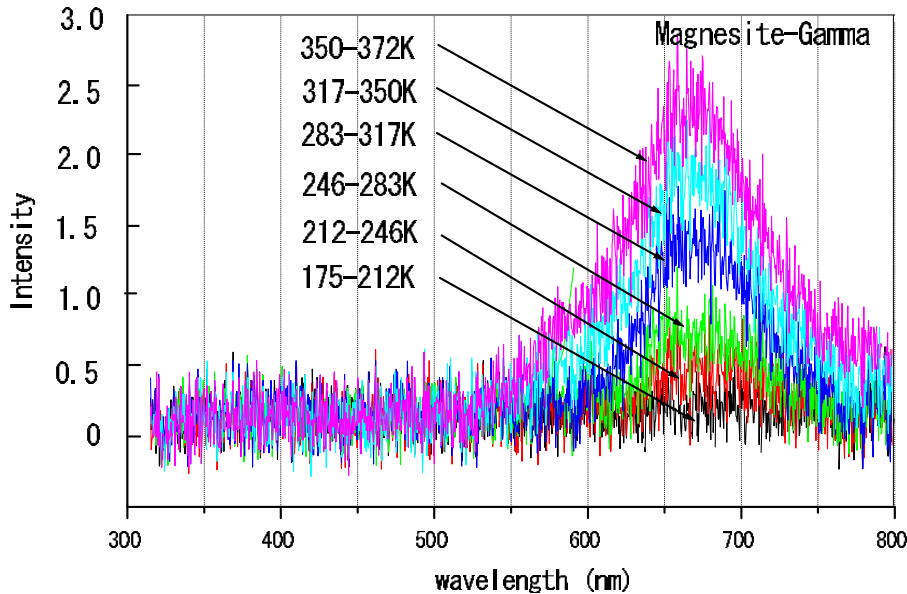


Fig. 6. Thermoluminescence spectra of magnesite (MgCO_3). The peak is at about 660 nm.

We have measured the thermoluminescence spectra of these samples using a spectrophotometer (including a CCD camera, Princeton Instruments, Inc.). The sample is put on the thermally-isolated plate, which has previously been cooled to liquid nitrogen temperature. The luminescence emission during warming is introduced to the CCD measuring system using an optically transparent fiber. The color of forsterite changed to dark-white and -gray (rather dark red-violet) after gamma-ray irradiation. The time of warming the samples from liquid nitrogen temperature to room temperature (333 K) is about 15 min. It should be noted that this time length is sufficient to observe the thermoluminescence, that is, photon is emitted at the thermal equilibrium state.

Fig.1 shows the luminescence spectra of forsterite (Mg_2SiO_4) after Gamma-ray irradiation. It shows a rather intense broad peak at about 645–655 nm (and a weak broad peak at 400–440 nm), and a sharp peak at 610 nm. The sharp peak at 610 nm appears as the sample warms from 200 K to room temperature. Other weak peaks at 590, 595, and 705 nm appear distinctly at above 200 K. The broad peak at 645–655 nm becomes suddenly strongest at 272 – 307 K, and the weak peaks fade into background. Above 307 K, the luminescence becomes weak gradually and another peaks at 595, 605, 705, 710 nm become prominent. The previous preliminary data of another sample of irradiated forsterite, its weight about 244 mg, have shown almost the same luminescence spectra, but it seems that the earliest luminescence starts at somewhat low temperature. Fig.2 shows the total strength of luminescence in Fig.1. A magnification of the emitted thermoluminescences spectrum of the most low-temperature at the range about 164–200 K in Fig.1 is shown in Fig.3.

In contrast, the luminescence is scarcely visible in natural olivine (from Egypt, Fo_{90}). As for enstatite, we have detect the luminescence spectra, which is shown in Fig.4. Two broad peaks appear at about 545 and 760 nm, and

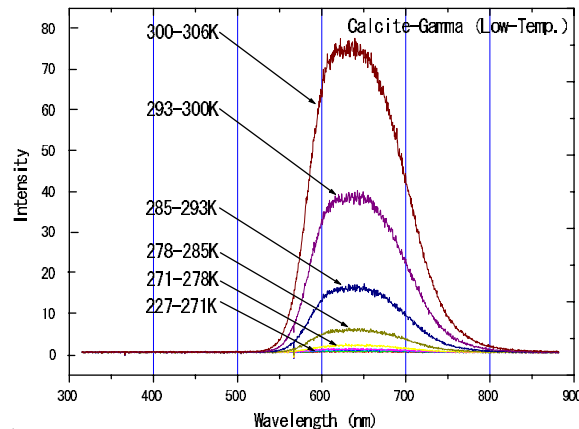


Fig. 7. Typical thermoluminescence spectrum of calcite (CaCO_3) at 227–300 K. The peak of CaCO_3 is at about 640 nm.

these peaks are very weak compared with the spectra of forsterite. The weight of enstatite used was 114 mg, while that of forsterite was 138 mg as mentioned before.

Fig.6 shows the measurement of luminescence spectra of magnesite MgCO_3 after Gamma-ray irradiation. The spectrum of MgCO_3 shows a very broad peak at about 660 nm, and this peak becomes strong at 350 – 375 K. The irradiated magnesite is a bulk sample (about $5 \times 5 \times 2$ mm size) and its weight is 93 mg. It should be emphasized that the thermoluminescence of MgCO_3 also has sufficient strength to be visible easily. The color of magnesite changed to a rather white-gray with pink-yellow tendency after gamma-ray irradiation.

We have also measured the thermo-dependence of luminescence of calcite. The present luminescence of calcite is shown in Fig.7 and Fig.8. The intensity becomes stronger as temperature increased from 77 K, and the peak position is 620–640 nm at 300 K. Above 300 K, the intensity became further stronger, and at 327 – 337 K the inten-

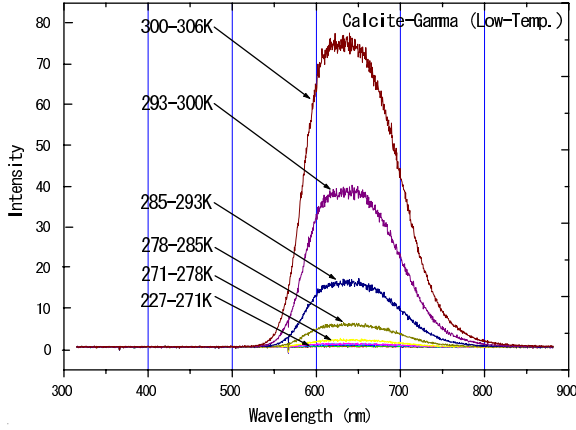


Fig. 8. Typical thermoluminescence spectrum of calcite (CaCO_3) at 300-337K. The peak shifts toward to 600nm.

sity became strongest, the shape of luminescence changed and the peak position shifts to around 605 nm. Higher temperature than 337 K, the intensity became weak.

3. Related problems and the other measurements

We also investigated the effect of irradiation on both a bulk sample and particle samples embedded in polyethylene using neutrons to a dose of $8 \times 10^{16} n_f/\text{cm}^2$ at 10 K and 70 K using the low-temperature irradiation facility of Kyoto University Reactor (KUR-LTL: Low Temperature Loop)(Okada et al., 2001). After neutron irradiations of 75 hour, samples are stored in liquid nitrogen for several months to wait a decay of radioactivity.

We measured the absorption coefficient of a neutron-irradiated forsterite and enstatite particles using a FTIR spectrometer(Nicolet, Nexus 670) over the wavelength range from 30 – 100 μm . The samples were irradiated at 70 K using a neutrons in September, 2000. The gross feature of spectrum show no apparent difference from the spectrum (Koike C. et al. 2000, 2000) taken before irradiation. One of the reasons for this result may be that the fast neutrons strongly collide with hydrogen atoms in the polyethylene and only weakly collide with sample particles dispersed in polyethylene.

We have also measured the thermoluminescence spectrum of neutron-irradiated bulk sample of forsterite, which is shown in Fig. 5. At low-temperature, two peaks appeared at about 450 and 650 nm, but above 253K one peak at 450nm became weak. The peak at 650nm is almost the same as Gamma-ray irradiated case, however, the strength of luminescence is considerably weak. This reason is not known yet.

For future irradiation experiments using a stronger beam of neutrons in the center of the reactor, we are interested in an accurate measurement of the impurity component in our forsterite sample, because impurity component with long a half period brings serious difficulties in measurements of irradiated samples. Previously, Takei and Kobayashi (1974) reported that the spectrographic anal-

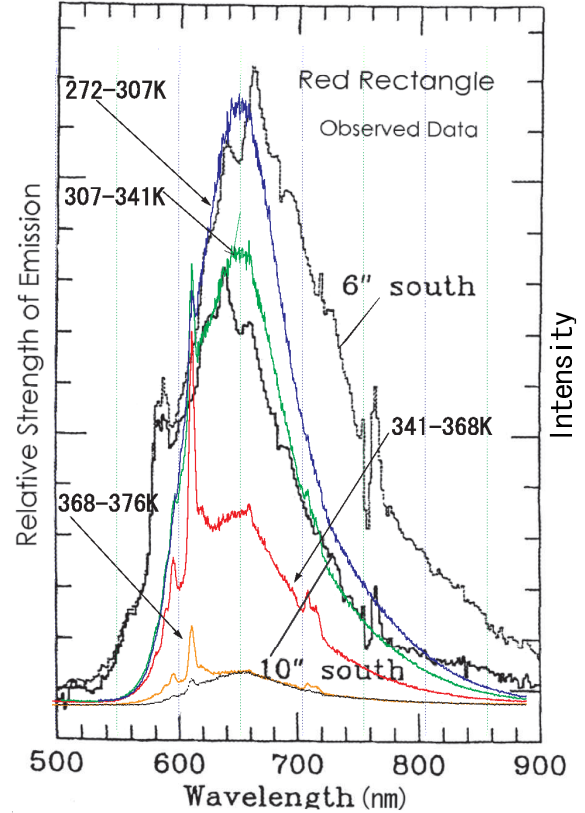


Fig. 9. Comparison with Observation Data of Red Rectangle Spectrum (From Witt A.N. and Boroson T.A. 1990) and Forsterite Spectrum (the right-side of Fig.1)

ysis shows that the forsterite sample is pure, but using neutron activation analysis, an infinitesimal quantity of Ir at about 16–18 wt ppm had been found. This level of Ir impurity is below the detection limit of the spectrographic analysis. We have also measured our sample using radio activation analysis, and confirmed that our sample is almost pure; that is, other elements except for Mg, Si, O and an infinitesimal quantity of Ir are not detected.

4. Similarity to ERE of Red Rectangle Spectrum

It is remarkable fact that the observed spectrum of ERE (Extended Red Emission) of the Red Rectangle (Witt and Boroson 1990) given in Fig. 9 shows characteristic feature. As for carriers of ERE, possible size effect of crystalline silicon nanoparticles has been discussed (Ledoux G. et al. 2001). It has been shown that the photoluminescence spectra of these nanoparticles can explain the gross structure of ERE spectra, such as the peak position and the full width at half maximum of ERE spectra etc. However, evidence of existence of crystalline silicon has not yet been observed. (Li and Draine 2001,2001)

We have interested that the luminescence of forsterite is very similar to ERE of Red Rectangle (peak at 652-684 nm, half width 70-90 nm) among many objects. Generally, ERE from interstellar dust consists of a broad, featureless emission band peaking at 610-820 nm, and width

60-100 nm. Among many objects, Red Rectangle show strong emission of one order stronger and exhibits the characteristic feature. Furthermore, crystalline silicates such as forsterite and PAH dust had been founded in Red Rectangle by ISO observations. The present irradiated forsterite also shows very strong luminescence (peak at 640-660 nm) at about 280-308 K and half width of about 100 nm, and characteristic peaks at 590, 595, and 705nm.

It should be also remembered that forsterite and enstatite have been found by many ISO observations in many oxygen rich young and evolved stars. However, ERE is observed in many carbon rich stars. It is important to note that Red Rectangle is very interesting object with both carbon and oxygen rich characteristics, that is, both forsterite and PAH were found. It should be emphasized that our thermoluminescence spectrum of forsterite at about 645–655 nm and at 590 nm is very similar to the ERE of the Red Rectangle.

5. Discussion

For the irradiation circumstance, little is known around ERE object. However, it should be noted that interstellar and circumstellar space is typically at extremely low temperature and is always irradiated by electromagnetic radiation and by cosmic ray particles over cosmological time-scale. Furthermore, it is well known that extremely large fluxes of neutrons and gamma-rays have been emitted during super-nova explosions. The effect of this radiation will accumulate in the low temperature environment. It will only be observed provided that the condition to release the accumulated energy is realized in circumstellar space. This may be occur when irradiated dust move to a warmer domain in interstellar or circumstellar environment.

The accumulated energy in irradiated matter is released by thermoluminescence when the matter is warmed. The rate of this release is dominated by Boltzman factor, and in extremely low temperature its rate is practically infinite. In connection with this problem, it is interesting fact that the thermoluminescence spectrum of particles in Tyrrhenian Sea exhibits the typical peak at the layer of year 1054, 1006, 1181 etc., corresponding to the year of super-nova explosions (Castagnoli G. C. et al., 1982). In their estimate, the energy emitted during super-nova explosions of Crab nebula at year 1054 is about 10^{47} J and the estimated flux of energy at the point of distance of the radius of the Galaxy is about 10^5 J/m².

Though most energies of explosions are carried by neutrinos, a certain rate will be carried by X and gamma rays. It will be possible in extremely low-temperature environment such as interstellar or circumstellar space, the effects of irradiation are "frozen" and almost stable in extremely long time, and various irradiation effects, such as often observed super-nova explosions or always irradiated cosmic rays, are accumulated.

The irradiation on matter generally cause lattice defects of crystal, and a certain kind of this effect is observed in thermoluminescence. It is known in many experiences that the irradiation effect on solid can be erased by annealing it to several hundred degrees. In other word, the effects of irradiation remains in a certain kind of form below this temperature and it may be observed still in effects different from the thermoluminescence.

6. Summary

In this paper, we report the data for gamma-ray irradiation of some simulated interstellar materials. Forsterite (Mg_2SiO_4), enstatite (MgSiO_3) and magnesite (MgCO_3) exhibit interesting thermoluminescences spectra.

We have emphasized that the thermoluminescence of forsterite appears at low temperature at about 160-200K. In particular, it should be emphasized that the spectrum of forsterite at about 645–655 nm and at 590 nm is very similar to the ERE of the Red Rectangle. (Witt and Boroson 1990) It should be noted that possible size effect of crystalline silicon nanoparticles can explain the gross structure of ERE spectra (Ledoux G. et al. 2001). However, this model can explain only the gross structure of spectrum of ERE, and seems not to explain the characteristic features of 590nm. The most interesting fact of size effect is the peaks of gross structure of luminescence shifts depending on the size of nanoparticles. It is interesting problem to examine possible existence of similar effect in forsterite nanoparticles.

Finally, the irradiation on matter is expected to cause various effects on matter in addition to thermoluminescence effects. Investigating these effects in the context of astrophysics is further problem.

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